Response to Referee#1:

We would like to thank the reviewer for the careful evaluations and positive comments on our paper, which improved the paper so much. We have revised the manuscript according to the reviewer's detailed comments. Please find the responses to the reviewers.

Reviewer's comments are in plain face.

Author responses are in blue color.

Changes in the manuscript are in red color.

Comments to the Author:

It is a pleasure to review the manuscript "Properties of aerosols and formation mechanisms over southern China during the monsoon season" by Chen et al. This manuscript addresses an important science question: what are the characteristics of size distribution and formation of atmospheric aerosols in southern China and how they are affected by local and long-range transport? Given the heavy PM concentrations in that region, answering this question has practical implications for public health. This study makes diligent use of a unique in-situ dataset, modeling, and remote-sensing products, investigates various physical and chemical mechanisms of aerosol (including secondary) formation and evolution, and provides some new insights into this scientific issue. The paper is comprehensive in its scope, well organized and well written, and the research quality is high. I suggest to accepting this manuscript after the authors clarify the following points, which are mostly minor concerns and editorial changes:

- Line 150, the model required the use of measured: did you have all those measurements from your site observations?

Response:

Yes, all of these required parameters were observed at these three sites, including ambient temperature, relative humidity, and the concentration of sulfate, nitrate and ammonium. We have added more information about chemical analysis in line 151-156:

'The mass concentrations of six cations (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, and Ca^{2+}) and seven anions (F^-, Cl^-, NO_2^-, Br^-, SO_4^{2-}, NO_3^- and PO_4^-) were analyzed using an ion chromatography (ICS-3000, DIONEX. Thermal Optical Transmittance (TOT) technique was employed to analyze the quartz filter samples to determine the mass concentrations of organic carbon (OC) and elemental carbon (EC) by the use of Sunset Laboratory OCEC Carbon Aerosol Analyzer.'

- Line 180, use of GDAS: why did you use higher resolution WRF/Chem

meteorological fields in place of GDAS?

Response:

WRF model could provide higher resolution meteorological data, further to improve the dispersion simulations and lead to an overall better simulation (Stohl, 1998). In addition, a novel convective scheme has been added in the FLEXPART –WRF (Brioude et al., 2013), which also could improve the model simulation, especially for finer scale applications. So meteorological fields provided by WRF instead of GDAS were used in this study. We have added an explanation in line 205-209:

‘ The application of FLEXPART –WRF with a novel convective scheme being added improves the dispersion simulations and results in an overall better simulation, especially for finer scale applications (Brioude et al., 2013; Stohl, 1998). In this case, the region was modeled with a spatial resolution of 27×27 km and a temporal resolution of 1 hour.’

- Line 187: the sentence is confusing. It looks like you use WRF simulated fields as input to FLEXPART, which contradicts with the previous statement of using GDAS?

Response:

Just as the reviewer said, WRF simulation provided meteorological fields for ELEXPAET, while GDAS was applied by HYSPLIT. So we have deleted this sentence and just kept the previous statement in line 201-205:

‘HYSPLIT uses single air parcels to compute trajectories with the use of Global Data Assimilation System (GDAS, 1°×1°) as input data. FLEXPART, on the other hand, uses a larger number of air parcels to compute trajectories based on the meteorological predictions provided by mesoscale model WRF.’

- Line 200: Which two sites are referring to here? There are three sites listed in Table 1.

Response:

We are sorry for our imprecise statement. Actually the range of [0.52, 0.55] and [0.72, 0.76] is the range for three sites not only for two sites. We have re-written this part to avoid confusion in line 226-228:

‘In terms of the mass size distribution, the percentage of $PM_{1.0}$ to PM_{10} was 60.2%, 66.3% and 75.0%, and $PM_{2.5}$ to PM_{10} was 88.0%, 92.6%, 91.7% in GZ, ZH and JFM, respectively.’

- Line 203-208: very nice analysis!

Response: Thanks so much for the review’s acceptance.

- Line 287: something is missing the sentence “far-upwind urbanization and biomass burning”. Urbanization cannot be transported.

Response:

Thanks for the reviewer’s reminder, we have added more information in line 316-319:

‘We investigated these days and find that emissions that long-range transported from far-upwind areas with highly urbanization or with the existence of biomass burning are responsible, as discussed later in this paper.’

- Line 304,”the sites”:which sites? - The quality of some figures are acceptable but not very good. For instance: o Figs 3c&d: the figure labels need to be improved. It is hard to read. o Fig 4:again the x-axis label is hard to read. o Fig 6 labels need to be improved.

Response:

Thanks for pointing out this issue. The sites represent the three sites. We have clarified it in line 339-341:

‘Additionally, it was determined that during these times at the three sites there was an abnormally high amount of low cloud cover 60-70% and a relatively higher relative humidity (75~83%) (Table 2).’

Thanks so much for the review’s suggestion, we have improved and re-plotted the figures in the main text.

- Line 417:a nice example of using backward trajectories to test your hypothesis!

Response: Thanks so much for the review’s acceptance.

- Section 3.6: can authors elaborate on the MODIS Fire products? Are these daily products or twice a day or 8-day?

Response:

Thanks for the reviewer’s suggestion, the MODIS fire products are daily products, we have provided some information about MODIS Fire products in the section ***2 Measurements and methodology*** in line 184-193:

‘Aerosol optical depth (AOD), fire products including Fire Radiative Power (FRP), and Fire Quality Assurance [QA] data, were obtained from the MODIS sensors aboard both the AQUA and TERRA satellites. Specifically, we obtained the Collection 6, 3km Level 2 swath product for AOD (Remer et al., 2013), and Collection 5.1, 1km Level 2 swath products for FRP and QA (Giglio et al., 2006). The Collection 5.1 active fire products are daily products and have been improved based on the previous collection 5.0 products. All of the data are cloud-screened, with AOD data being computed using different algorithms over land and water, and the fire data using 19

different channels for quality assurance. We only accept values for FRP and Fire Count where the QA is at least 90%.'

And in line 550-555:

'MODIS fire hotspots are not very useful in wet and tropical regions. Since MODIS fire hotspots are obstructed by both clouds and high levels of aerosols in the atmosphere, both of which are found associated with tropical forest fires. Additionally, due to the highly wet ground surface, a significant amount of the fires may low temperature and therefore not observable using the MODIS sensors (Cohen, 2014, Giglio et al., 2006; Yu et al., 2015)'

Editorial suggestions:

- Line 92: replace "The data is" by "those data were"

Response:

Thanks and we have modified it.

- Line 94: insert "of size distribution" between "characteristics" and "in"

Response:

Thanks and we have added it

- Lin 98-99: something is missing in "and the impacts mixing sea-salt and urban pollutants"

Response:

Thanks for reminding and we have added more information in line 106-107.

'and the impacts mixing sea-salt and urban pollutants on the characteristics of size distributioin .'

- Line 103: it would be good to show photos of these sites to give an idea of site environments

Response:

Thanks for the suggestion, we have added photos of these sites in Fig. 1

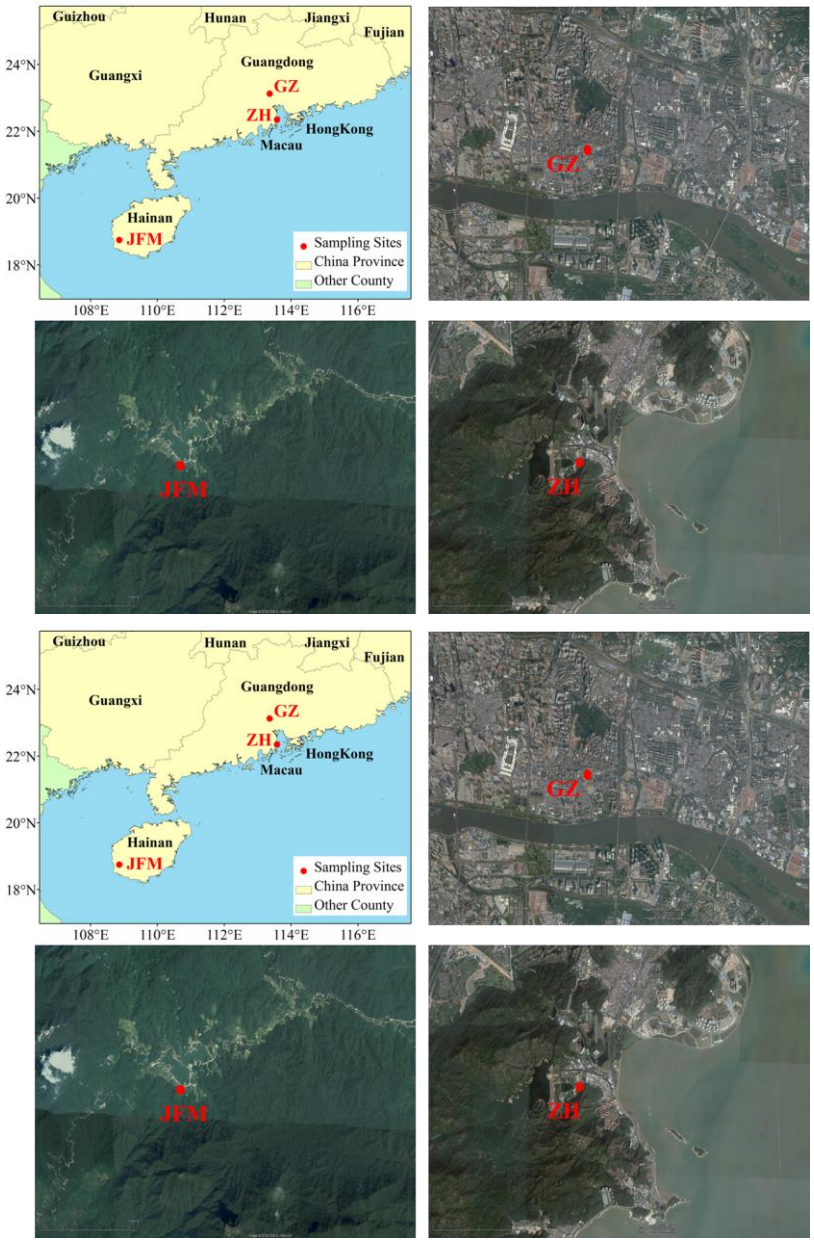

Figure 1. Location of sampling sites in Southern China: GZ (Guangzhou), ZH (Zhuhai), and JFM (Jianfeng Mountain) and their surrounding environments

- Line 110: replace "The first site was set at (23.12N, 113.36E), on" by "The first site (23.12N, 113.36E), is located on"

Response:

Thanks and we have modified it.

- Line 111: replace "an urban mega-city" by "a mega-city"

Response:

Thanks and we have modified it.

- Line 149: delete “to”

Response:

Thanks and we have modified it.

- Line 170: replace “All of the data is” by “All data are”

Response:

Thanks and we have modified it.

- Line 194: delete“mass”

Response:

Thanks and we have modified it.

- Line 212-213: replace “no matter what the particle size was” by “irrespective of particle size”

Response:

Thanks and we have modified it.

- Line 217: should “formed” be “from”

Response:

Thanks and we have modified it.

- Line 219: delete “with”

Response:

Thanks and we have modified it.

– Line 226: delete “of”

Response:

Thanks and we have modified it.

- Line 242: replace “showing” by “shows”

Response:

Thanks and we have modified it.

- Line 281: replace “high levels” by “high-level”

Response:

Thanks and we have modified it.

- Line 288: replace “as talked about later” by “as discussed later in this paper”

Response:

Thanks and we have modified it.

- Line 303: replace “show” by “shown”

Response:

Thanks and we have modified it.

- Line 312: replace “evidence” by “evidenced”

Response:

Thanks and we have modified it.

- Line 323: replace “of the” by “shows that”

Response:

Thanks and we have modified it.

- Line 489: delete the second “instead”

Response:

Thanks and we have modified it.

- Line 527-529: replace “Based on specific case studies, some models of the air flow, and remote sensing, the impacts of chemistry and atmospheric transport were investigated” by “These site observations, together with model simulations and Remote-sensing data, were used to investigate impacts of chemistry and atmospheric transport”.

Response:

Thanks and we have modified it.

- Line 547: delete “further”

Response:

Thanks and we have modified it.

- Line 554: be specific on “they”. What are they?

Response:

Thanks and ‘they’ represent ‘OC and EC’, we have clarified it.

Reference:

Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., Angevine, W., Evan, S., Dingwell, A., Fast, J.D., Easter, R.C., Pisso, I., Burkhart, J. and Wotawa, G.: The Lagrangian particle dispersion model FLEXPART-WRF version 3.1, Geoscientific Model Development, 6, 1889-1904, doi:10.5194/gmd-6-1889-2013, 2013.

Stohl, A.: Validation of the lagrangian particle dispersion model FLEXPART against large-scale tracer experiment data, Atmospheric Environment, 32, 4245-4264, doi:10.1016/S1352-2310(98)00184-8, 1998.

Yu, C., Chen, L.F., Li, S.S., Tao, J.H. and Su, L.: Estimating Biomass Burned Areas from Multispectral Dataset Detected by Multiple-Satellite, Spectroscopy and Spectral Analysis, 35, 739-745, doi:10.3964/j.issn.1000-0593(2015)03-0739-07, 2015.